Screening in crystalline liquids protects energetic carriers in hybrid perovskites

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Hybrid lead halide perovskites exhibit carrier properties that resemble those of pristine nonpolar semiconductors despite static and dynamic disorder, but how carriers are protected from efficient scattering with charged defects and optical phonons is unknown. Here, we reveal the carrier protection mechanism by comparing three single-crystal lead bromide perovskites: CH3NH3PbBr3, CH(NH2)2PbBr3, and CsPbBr3. We observed hot fluorescence emission from energetic carriers with ~102-picosecond lifetimes in CH3NH3PbBr3 or CH(NH2)2PbBr3, but not in CsPbBr3. The hot fluorescence is correlated with liquid-like molecular reorientational motions, suggesting that dynamic screening protects energetic carriers via solvation or large polaron formation on time scales competitive with that of ultrafast cooling. Similar protections likely exist for band-edge carriers. The long-lived energetic carriers can enable hot-carrier solar cells with efficiencies exceeding the Shockley-Queisser limit.

Hybrid perovskites (HOIPs) with the general formula A3PbX5 (X = I, Br, or Cl), where A+ is an organic cation such as methylammonium (MA), CH3NH3+, or formamidinium (FA), CH(NH2)2+ (I–4), are usually processed from solutions at room temperature for solar device fabrication. A high defect density is unavoidable (5), yet these defective semiconductors exhibit carrier properties expected of defect-free and nonpolar inorganic semiconductors. These properties include (i) inverse temperature dependence of charge carrier mobility (μ) with a power law, μ ∝ T−3/2 (6–8), predicted for coherent transport hindered by acoustic phonon scattering (9); (ii) long carrier diffusion length (up to 102 μm) and carrier lifetime (~1 μs), which are unexpected for a semiconductor with modest charge carrier mobility (μ ∼ 1 to 100 cm2 V−1 s−1) (2–4, 10); and (iii) low electron-hole recombination rate constant (~10−10 cm3 s−1) that rivals those of the purest crystalline semiconductors (e.g., GaAs) (6, 11, 12). All of these properties suggest that charge carriers in HOIPs are protected from scattering with charged defects, optical phonons, and each other. Based on the presence of extensive dynamic disorder on multiple time scales (13–16), Zhu and Podzorov proposed that formation of large polarons (17) may explain the protection of charge carriers in HOIPs (18).

To understand the carrier protection mechanism in HOIPs, we have compared carrier and structural dynamics in single-crystal lead bromide perovskites with three different cations: methylammonium (MAPbBr3), formamidinium (FAPbBr3), and cesium (CsPbBr3) using time-resolved photoluminescence (TR-PL) and time-resolved optical Kerr effect (TR-OKE) (19) spectroscopies. These bulk-sensitive measurements on pristine single crystals provide insights into intrinsic carrier and structural properties. Using TR-PL, we show long-lived energetic carriers (electronic temperature ≥1000 K) that cool down with markedly long time constants of ~102 ps in MAPbBr3 or FaPbBr3, but not in CsPbBr3. In TR-OKE measurements, we observed liquid-like reorientation motions on subpicosecond to picosecond time scales in MAPbBr3 or

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**Fig. 1. Time-dependent PL spectra revealing long-lived hot carriers.** (A) SEM image and selected area electron diffraction pattern (inset) of a single-crystal MAPbBr3 microplate. Scale bar: 10 μm. (B) Pseudo-color (intensity) plot of TR-PL spectra from a single-crystal MAPbBr3 microplate at room temperature under 3.08-eV excitation and an excitation density of 1.7 μJ cm−2. (C) PL intensity decay kinetics at 2.3 eV (red) and 2.6 eV (blue) and single exponential fits (solid). The PL intensity at 2.6 eV has been multiplied by a factor of 1500. (D) PL spectra (squares) at indicated delay times and fits (colored curves) to the two-temperature model. The black solid and dashed curves show components in the fit to the spectrum at 50 ps. (E) Extracted electronic temperature from the hot-carrier distribution as a function of delay time. The data points are average values from three independent microplate samples. The red curve is a single exponential fit to the first 0.5 ns, which gives a hot-carrier relaxation time constant of 150 ± 30 ps.
FAPbBr$_3$, but not in CsPbBr$_3$. The correlation between TR-PL and TR-OKE measurements suggests that the liquid-like reorientational motions of organic cations provides protection for energetic charge carriers in HOIPs via solvation and screening on sufficiently fast time scales to compete with carrier cooling by optical phonon scattering. Further corroborating this, we observed the absence of hot fluorescence emission in the low-temperature orthorhombic phase of MAPbBr$_3$, where the liquid-like motions of MA ions are frozen. Thus, an HOIP possesses both crystalline solid and liquid-like behaviors and belongs to the family of solids called plastic crystals ($20, 21$). Such an efficient screening process, which is similar to large polaron formation in physics ($17$) or solvation in chemistry, likely also exists for band-edge carriers whose long lifetime (up to microseconds) ensures coupling to the slower dynamics (>1 ps) of the soft PbX$_3$ sublattice.

We synthesized optically flat, single-crystal APbBr$_3$ ($A = MA, FA$, or Cs) samples in the form of either macroscopic (~1 mm in lateral dimensions and ~hundreds of micrometers in thickness) ($10, 22$) or microscopic plates (~tens of micrometers in lateral dimensions and ~hundreds of nanometers in thickness) ($23–25$), as detailed in the supplementary materials ($26$). The optical and scanning electron microscope (SEM) images of the single crystals are shown in figs. S1 and S2 ($26$). The corresponding powder and single-crystal x-ray diffraction (XRD) data confirmed that the liquid-like reorientational motions of the single crystals are shown in figs. S1 and S2 ($26$). The corresponding powder and single-crystal x-ray diffraction (XRD) data confirmed single crystallinity. All samples were stored in a nitrogen glove-box after growth and transferred to vacuum cryostats for spectroscopic measurements at a base pressure ≤10$^{-3}$ torr. We conducted TR-PL measurements using a home-built inverted microscope and time-correlated single photon counting (TCSPC). The TR-PL results from microplates are shown below and, along with those from macrocrystals, in the supplementary materials ($26$). Optically thin crystals in the former minimized the effects of carrier diffusion into the bulk and to reabsorption of PL emission ($27, 28$). The TR-OKE measurements were carried out on macrocrystals with both pump and probe photon energies below the band gap.

A pseudocolor plot of TR-PL spectra from a single MAPbBr$_3$ microplate excited at 3.08 eV and at a temperature $T = 293$ K shows the presence of hot PL emission on the 10$^3$-ps time scale (Fig. 1B); see fig. S5 for results from additional MAPbBr$_3$ crystals ($26$). At this temperature, MAPbBr$_3$ is in the cubic phase, where the MA cation can freely rotate in the cubic cell of PbBr$_6^2−$ cage ($13−16$). Photocexcitation in MAPbBr$_3$ at room temperature and at the exciton densities used (≤7 × 10$^{19}$/cm$^3$) gives primarily uncorrelated charge carriers instead of excitons ($29$), as verified by the quadratic dependence of PL intensity on laser pulse energy (fig. S3). The PL spectra feature strong band-edge emission at 2.31 eV and, at short times (<1 ns), a broad high-energy tail extending more than 300 meV above the band edge, which is attributed to radiative recombination of hot carriers. As shown by a comparison of PL decay kinetics at low (2.3 eV) and high (2.6 eV) energies in Fig. 1C, within 1 ns, the band-edge emission remains almost constant, but hot-carrier luminescence decays with a time constant of $\tau = 160 ± 10$ ps. For $t > 500$ ps, the two kinetic curves merge and show identical PL decay kinetics with a time constant of $\tau = 4.4 ± 0.1$ ns.

To analyze the hot PL emission, we assumed that the PL spectrum at each delay time is a sum of radiative recombination from thermalized carriers, $T_e = 293$ K, and that from a hot distribution characterized by an effective carrier temperature $T^*$. The functional form for each distribution is given by the product of the combined density of state (DOS) distribution in the parabolic band approximation for the conduction and valence bands and the Fermi-Dirac function ($26$). We carried out the fitting globally using PL spectra at all delay times, and Fig. 1D shows fits (solid colored curves) along with data (squares) at selected delay times; also shown are the cold (black solid) and hot (black dashed) components of the fit to the PL spectrum at 50 ps. In the supplementary materials ($26$), we show a complete set of global fitting results (fig. S4).

The relative amplitude of PL from hot carriers, $\gamma = 33 ± 5\%$, remained constant, but the effective temperature $T^*$ decreased with time, consistent with the cooling of hot carriers. As shown in Fig. 1E, the hot-carrier temperature started at $T^* = 1250 ± 200$ K and cooled down with a cooling time constant of $\tau_c = 150 ± 30$ ps. Beyond ~0.5 ns, cooling became very slow and $T^*$ approached the asymptotic value of $T^*_{eq} = 670 ± 100$ K. Note that $T^*_{eq}$ was higher than the equilibrium sample temperature of 293 K. This phenomenological $T^*_{eq}$ resulted from PL spectral broadening, which has been assigned to the phonon dressing effect for band-edge emission ($30$). Thus, the actual range of cooling is $\Delta T^* = 550$ K within ~1 ns. The above analysis should be viewed as a qualitative estimate because of the complexity of cooling dynamics, the spectral broadening, and the simplicity of the two-temperature model. The model might underestimate the hot-carrier distribution because (i) radiative recombination rates from energetic carriers are lower than that from band-edge carriers ($31$) and (ii) hot fluorescence emission is reduced as compared to band-edge emission, owing to preferential reabsorption of the former ($27, 28$).

We compare carrier cooling dynamics in MAPbBr$_3$ with those in hybrid FAPbBr$_3$ and the all-inorganic CsPbBr$_3$ at room temperature. Similar to MAPbBr$_3$, photoexcitation of FAPbBr$_3$ or CsPbBr$_3$ at room temperature also gave uncorrelated charge carriers at an excitation density of ~7 × 10$^{19}$/cm$^3$ (fig. S3). The FAPbBr$_3$ crystal contained freely rotating molecular cations (FA$^+$) in the room-temperature cubic phase, albeit with a dipole moment less than that of MA$^+$ ($32$). In contrast, such reorientational motion was absent in CsPbBr$_3$, which is in the orthorhombic phase at room temperature. The pseudocolor plot of TR-PL spectra in the first 500 ps and selected PL spectra at indicated delay times are shown in Fig. 2. A and C, for FAPbBr$_3$ and in Fig. 2, B and D, for CsPbBr$_3$ microplates. TR-PL spectra for additional FAPbBr$_3$ and CsPbBr$_3$...
single-crystal microplates are shown in figs. S6 and S7, respectively. Similar to MAPbBr3, we observed long-lived energetic carriers in FAPbBr3 (Fig. 2, A and C). Analysis with the two-temperature model gave a relative hot PL population of γ = 23 ± 3% (fig. S8). The effective hot-carrier temperature was ~1400 K at 50 ps and cooled down with a time constant of 190 ± 20 ps within ~0.5 ns. The cooling slowed drastically at t ≥ 0.5 ns and, at 10 ns, reached a value of ~680 K that we attributed to PL spectral broadening. In contrast to FAPbBr3 or MAPbBr3, we observed no spectral shape evolution with time for CsPbBr3 (Fig. 2, B and D), indicating that all excited carriers had relaxed to the band edge within our time resolution (~20 ps). We also confirmed the presence of long-lived hot carriers in MAPbBr3 and FAPbBr3, but not in CsPbBr3, using millimeter-sized single crystals (fig. S9).

A key difference between the HOIPs (MAPbBr3 and FAPbBr3) and the all-inorganic CsPbBr3 is the presence of reorientational motion of dipolar molecular cations at room temperature in the former, but not in the latter. To quantify the liquid-like reorientational motion directly, we applied TR-OKE spectroscopy (39, 33). In this approach, a polarized optical field (pump pulse) induces anisotropy in the refractive index and, thus, transient birefringence in the sample. After a controlled time delay, a second optical field probes the decay of transient birefringence based on polarization rotation (supplementary materials).

This technique has been used to probe reorientational motions in liquids (29), in solid-liquid phase transitions (33), and in the plastic crystal succinonitrile (34). There are generally four types of responses in molecular liquids: (i) an instantaneous electronic response; (ii) an ultrafast response (<200 fs) caused by inertial reorientation (also called librations or frustrated rotation) when there is polarization anisotropy; (iii) an intermediate response (~400 to 600 fs) caused by local-interaction-induced anisotropy associated with the molecular reorientational motion; and (iv) a slow response (>1 ps) from the diffusive reorientation of a molecule with permanent polarizability anisotropy (10).

Figure 3 compares TR-OKE responses from CsPbBr3, MAPbBr3, and FAPbBr3. The signal from CsPbBr3 (Fig. 3A) was characterized by an instantaneous electronic response (i), which was identical to the pump-probe cross correlation (gray, 70-fs full width at half-maximum), and an ultrafast response with a lifetime of τ = 140 ± 10 fs, which we attributed to inertial reorientation when there is polarization anisotropy (29). For CsPbBr3, the polarization anisotropy came from local distortion of the unit cell and the inertial reorientation from libration of the distorted unit cell near a local potential minimum.

In contrast to CsPbBr3, the TR-OKE transients of either MAPbBr3 (Fig. 3B) or FAPbBr3 (Fig. 3C) were characterized by, in addition to the instantaneous (i) and ultrafast (ii) responses, prominent, long-time responses covering a broad time window, from ~10^-7 fs to ~2 ps. Indeed, both HOIP spectra are similar to typical TR-OKE responses from anisotropic molecular liquids (19, 33). As in liquids, we could assign the subpicosecond region (iii) and the ~1- to 2-ps region (iv) to rotational motions associated with local-interaction-induced anisotropy and to diffusive rotation of the molecular cations, respectively. The time scales of the corresponding motions are in good agreement with results from molecular dynamics simulations (13, 35), two-dimensional infrared spectroscopy (36), neutron scattering (13, 14), and nuclear magnetic resonance spectroscopy (16).

The TR-OKE transients in Fig. 3 reveal that molecular cations in MAPbBr3 and FAPbBr3 at room temperature behave like anisotropic molecular liquids, as in plastic crystals (33, 34). The liquid-like behavior in HOIPs is also supported by previous measurements on the real part of the dielectric permittivity, whose temperature dependence in the cubic and tetragonal perovskite phases is well described by the Kirkwood-Frohlich equation for polar liquids (15). The liquid-like behavior in MAPbBr3 or FAPbBr3, but not in CsPbBr3, correlates well with the presence of long-lived hot carriers in the former and suggests the role of molecular dipole motion in energetic carrier protection.

To further verify the critical role of liquid-like motion, we performed TR-PL measurements on MAPbBr3 single crystals at two lower temperatures, 180 and 77 K, corresponding to the tetragonal and orthorhombic phases, respectively (15, 16). Compared to the free rotation in the high-temperature cubic phase, the rotational motion of the MA cation is partially hindered in an anisotropic potential in the tetragonal phase (at 180 K) and completely frozen in the orthorhombic phase (at 77 K). The TR-PL spectra for MAPbBr3 crystals at 180 K are shown in Fig. 4, A and C; and revealed long-lived hot-carrier population in the tetragonal phase. Fitting to the two-temperature model yields a relative hot-carrier population of γ = 24%; the hot-carrier temperature started at ~1000 K and cooled down with a time constant of 150 ps (fig. S10), similar to results at 293 K in Fig. 1. In contrast, there was no hot luminescence at 77 K (Fig. 4, B and D), as shown by the absence of a high-energy tail or spectral shape evolution with time. Thus, energetic carriers must have relaxed to the band edge within our time resolution (~20 ps) in the orthorhombic phase. In conventional semiconductors, the opposite trend is observed: The hot electron lifetime increases with decreasing temperature (37).

The results presented here for MAPbBr3 and FAPbBr3 single crystals establish the presence of long-lived hot carriers with electronic temperature ≥1000 K and with ~10^-7 ps lifetimes in HOIPs. Such long-lived energetic carriers are unexpected for polar semiconductor materials, where efficient scattering of carriers (electrons and holes) with longitudinal optical (LO) phonons resulted in lifetimes on the order of 10^-6 fs for energetic carriers with ~10^2 meV excess energy (37). In a conventional model, long-lived energetic carriers become possible only (i) when the excess electronic energy is below that of the LO phonons and the much slower acoustic-phonon scattering becomes the dominant channel for electronic energy loss (37); or (ii) at high excitation densities, where hot phonons are not cooled fast enough and can heat the electronic degrees of freedom (38, 39). The former is important for excess electronic energy less than tens of milli–electron volts in most semiconductors. The latter is the well-known hot "phonon bottleneck," and...
the hot-carrier lifetime increases with excitation density, as was also observed recently for MAPbI₃ thin films at high excitation densities (≥10¹⁷ cm⁻³) (31, 40, 41). However, we observed energetic carriers (7° to 1000 K) with ~10⁻³ ps lifetimes for single-crystal HOIPs at excitation densities as low as 10⁹ cm⁻³ and, in contrast to the phonon bottleneck, the long-lived excess electronic energy actually decreased with increasing excitation density (fig. S11), as explained below in the large polaron model.

The correlation of the liquid-like motion with the presence of long-lived high fluorescence in HOIPs implies that the protection of energetic carriers comes from the molecular dipoles that screen their scattering with LO phonons. We argue that the ultrafast dynamics (tens of femtoseconds to 2 ps) of this screening process protects the energetic carriers and is responsible for the slower cooling (hundreds of picoseconds) later on. Within the theoretical framework of polaron physics (17) since the seminal work of Landau (42), a large polaron (a charge carrier dressed by long-range lattice deformation) must form from an electron or hole in the highly polarizable environment in a HOIP. However, unlike the conventional picture of a large polaron resulting from the Coulomb potential between an electron or hole and the ionic lattice, a charge carrier residing in the inorganic PbX₃ sublattice can additionally be screened by liquid-like molecular dipoles with reorientational freedom. Thus, large polaron formation in a HOIP bears a resemblance to solvation dynamics in solutions.

We can identify two possible scenarios from this liquid-like polarization environment: (1) Although a large polaron may form from the carrier-induced deformation of the lattice in either a HOIP or its all-inorganic counterpart, dynamic simulations suggest faster deformation dynamics in the former because of the coupling of the liquid-like motion of organic cations with the deformation of the PbX₃ framework (49). The faster dynamics in MAPbX₃ or FAPbX₃, compared to that in CsPbX₃, makes the former kinetic competition with ultrafast cooling via electron–LO phonon scattering. Although the TR-OKE transients in Fig. 3, B and C, show time scales for molecular reorientational motion on the order of ~10⁻² is to ~picoseconds, the collective motion responsible for initial large polaron formation in the presence of an extra carrier could occur at shorter time scales, as is well known in solvation dynamics of the hydrated electron (46). The presence of photoexcited carriers in a HOIP can also weaken the hydrogen bond interaction between an organic cation and the PbX₃ cage, leading to faster deformation dynamics (45). (ii) The dynamical organization of liquid-like molecular dipoles is expected to result in preformed local ferroelectric domains on nanometer scales (46, 47). A photogenerated charge carrier can move to a preformed domain or domain boundary on the order of femtoseconds (32), as determined by the inverse of the inter–unit-cell electronic coupling.

Although we do not know which of the two mechanisms above is more important, once a large polaron protection shield is formed around an electron or hole, the Coulomb interaction responsible for its scattering with LO phonons is screened, leading to a drastically reduced rate of hot-carrier cooling by orders of magnitude. Indeed, a recent four-wave mixing experiment comparing carrier dephasing time in MAPbI₃ and GaAs reported much reduced the many-body Coulomb interaction in the former (48). The large polaron model also explains the inverse dependence of long-lived excess electronic energy on excitation intensity (fig. S11). This observation is the opposite of what is expected from the conventional phonon bottleneck. Because of the competition for nuclear polarization, there is an effective interpolaron repulsive interaction that destabilizes the large polaron at high excitation densities (37). As a result of this destabilization of the large polaron protection shield, the cooling rate of an energetic carrier by LO-phonon scattering increases with excitation density, until the onset of a hot-phonon bottleneck at even higher densities (~10⁻⁶ cm⁻³) when the effective temperature of the more free-electron/hole-like carriers increases again with excitation density (31, 40, 41).

Because the long-lived excess electronic energy of the large polaron increases as excitation density decreases, this process may be relevant to the carrier density range under solar radiation and may lead to the realization of the elusive hot-carrier solar cell concept with a theoretical power conversion efficiency as high as 66% (49). In a conventional solar cell, excess energy from energetic carriers is lost to phonons before they can be harvested, and this loss is partially responsible for the Shockley–Queisser limit of ~31% in power conversion efficiency. The exceptionally long lifetimes (~10⁻⁵ ps) of energetic carriers in HOIPs may make hot-carrier harvesting possible.

REFERENCES AND NOTES
26. Supplementary materials are available on Science Online.
High-quality graphene via microwave reduction of solution-exfoliated graphene oxide

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Efficient exfoliation of graphite in solutions to obtain high-quality graphene flakes is desirable for printable electronics, catalysis, energy storage, and composites. Graphene oxide with large lateral dimensions has an exfoliation yield of ~100%, but it has not been possible to completely remove the oxygen functional groups so that the reduced form of graphene oxide (GO; reduced form: rGO) remains a highly disordered material. Here we report a simple, rapid method to reduce GO into pristine graphene using 1- to 2-second pulses of microwaves. The desirable structural properties are translated into mobility values of >1000 square centimeters per volt per second in field-effect transistors with microwave-reduced GO (MW-rGO) as the channel material and into particularly high activity for MW-rGO catalyst support toward oxygen evolution reactions.

Low yields of single-layered graphene, submicrometer lateral dimensions, and poor electronic properties remain as major challenges for solution-exfoliated graphene flakes (1–4). Oxidation of graphite and its subsequent exfoliation into monolayered graphene oxide (GO) with large lateral dimensions (5–7) produce an exfoliation yield of ~100%; however, despite numerous efforts, it has not been possible to completely remove the oxygen functional groups (1, 2, 8, 9) so that the reduced form of GO (rGO) remains a highly disordered material with properties that are generally far inferior to graphene grown by chemical vapor deposition (CVD graphene) (5). Although rGO has been widely demonstrated to be a potentially useful material for catalysis (10–13) and energy storage (14–18), even in its disordered form, efficient reduction of GO into high-quality graphene should lead to substantial enhancement in performance. Here we report a simple and rapid method to reduce GO into pristine graphene by using 1- to 2-second-long microwave pulses. The microwave-reduced GO (MW-rGO) exhibits pristine CVD graphene-like features in the Raman spectrum with sharp G and 2D peaks and a nearly absent D peak. X-ray photoelectron spectroscopy (XPS) and high-resolution transmission electron microscopy (HR-TEM) suggest a highly ordered structure in which oxygen functional groups are almost entirely removed. The desirable structural properties are translated into mobility values of >1000 cm² V⁻¹ s⁻¹ in field-effect transistors (FETs) with MW-rGO as the channel material and into exceptionally low Tafel slope values of ~38 mV per decade for MW-rGO catalyst support for oxygen evolution reaction (OER). These results suggest that reduction of GO using microwaves is highly efficient and realizes the goal of achieving high-quality graphene with desirable properties by solution exfoliation.

We used the modified Hummers’ method to oxidize graphite and solubilize it into monolayered GO flakes in water (19). The stable suspension of GO sheets in water allows them to be reconstituted in several different forms such as thin films (20), bucky paper (21), or fibers (22, 23). GO synthesized in this manner is electrically insulating because of the presence of oxygen functional groups that are covalently bonded with the carbon atoms (2). Substantial effort has been devoted to recover the conducting π states of sp²-bonded carbon atoms by removing oxygen functional groups via chemical (1, 24, 25) or thermal (26) reduction (even heating over 3000 K (23)). By carefully tuning the reduction procedure, it is possible to realize noteworthy optical (20, 27, 28) and electronic properties (27) of rGO that are substantially different from those of pristine graphene because the evolution of the oxygen functional groups during reduction is accompanied by the formation of defects in the graphene basal plane (29). Specifically, nanoscopic holes occur through loss of carbon as CO or CO₂ (30), and rearrangement of the carbon atoms in the graphene basal plane leads to formation of Stone-Wales types of defects (31). In addition, oxygen functional groups form highly stable ether and carbonyl groups (32) that are difficult to remove so that GO contains a residual oxygen concentration of 15 to 25 atomic % (32). These factors render rGO a highly defective material, with several studies reporting electronic conductivity and mobility values on the order of 1 cm² V⁻¹ s⁻¹ (33–35).

Flakes of GO with lateral dimensions as high as tens of micrometers are shown in Fig. 1A. We used microwaves from a conventional microwave oven operated at 1000 W for 1- to 2-s pulses to reduce GO [see the “Preparation of microwave-reduced graphene oxide (MW-rGO)” section in the supplementary materials (36)]. Irradiation of GO with microwaves has been reported previously (37–39), but the reduction efficiency has been low and the rGO remains highly disordered, as indicated by the presence of an intense and broad disorder D band and the absence of the 2D band in the Raman spectra. We irradiated GO...
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Science 353 (6306), 1409-1413.
DOI: 10.1126/science.aaf9570

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